Effects of transition metal substitution on the glass-formation ability and magnetic properties of Fe62Co9.5Nd3Dy0.5B25 glassy alloy

著者	張　偉
journal or publication title	Journal of Applied Physics
volume	91
number	8
page range	5227-5229
year	2002
URL	http://hdl.handle.net/10097/47360

doi: 10.1063/1.1457538

JOURNAL OF APPLIED PHYSICS VOLUME 91, NUMBER 8 15 APRIL 2002

Effects of transition metal substitution on the glass-formation ability and magnetic properties of $Fe_{62}Co_{9.5}Nd_3Dy_{0.5}B_{25}$ glassy alloy

Yi Long
School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, China

Wei Zhang[a)]
Inoue Superliquid Glass Project, Exploratory Research for Advanced Technology, Japan Science and Technology Corporation, Sendai 982-0807, Japan

Xinmin Wang and Akihisa Inoue
Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

(Received 24 September 2001; accepted for publication 11 January 2002)

Fe–Co–Nd–Dy–B glassy alloys are a current type of ferromagnetic material. To increase their glass-forming ability against the growth of a crystallization phase, the effects of the addition of transition metals TM (TM=V, Nb, Ta, Cr, Mo, and W) on the glass-formation ability and magnetic properties of $Fe_{62}Co_{9.5}Nd_3Dy_{0.5}B_{25}$ glassy alloy have been investigated. The substitution of 2 at. % elements TM (=Nb, Ta, Mo, and W) for Fe and Co increases crystallization temperature T_x and decreases the onset temperature of solidification T_m, leading to a significant increase in the thermal stability against crystallization for $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$. The difference ($\Delta T_x = T_x - T_g$) between T_x and the glass transition temperature T_g increases from 55 K at 0 at. % TM to 87 K at 2 at. % TM. The bulk glassy alloy with a diameter up to 1.2 mm was produced by copper mold casting. Also no distinct changes in T_g, T_x and ΔT_x are seen for the addition of Cr and V. The results can be explained by the difference of atomic size in the additional elements. The saturation magnetization decreases slightly by the addition of 2 at. % TM elements. The saturation magnetization and coercive force of glassy $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloys are 1.13 to 1.19 T and 3.85–4.98 A/m, respectively. [DOI: 10.1063/1.1457538]

I. INTRODUCTION

Ferromagnetic metallic glasses have attracted increasing interest because they have a wide supercooled liquid region ΔT_x before crystallization and high glass-formation ability against the growth of a crystallization phase.[1–2] The high glass-formation ability implies that the glassy alloys have a large viscous flowability in the supercooled liquid region ΔT_x. Therefore ferromagnetic metallic glasses with the high glass-formation ability are workable to various complicated shapes by utilizing the low viscosity and the large flowability.

It is well known that the glassy alloys with high glass-formation ability satisfy the following three empirical rules:[3] i.e., (1) multicomponent systems consisting of more than three elements, (2) significant difference in atomic size above about 12% among the main three constituent elements, and (3) large negative heats of mixing among the elements. Very recently, based on the empirical rules, a current type of ferromagnetic metallic glass Fe–Co–Ln–B (Ln=rare earth elements) with a wide supercooled liquid region was prepared.[2–4] The Fe–Co–Ln–B metallic glasses exhibit good soft magnetism with saturated magnetization of 0.84–1.66 T and coercivity (H_c) of 5.0–36 A/m.[4,5] It is found that the glassy phase of Fe–Co–Ln–B alloys consists of (Fe, Co)–B-based rigid local structure units and the Ln atom is inserted between them. Lanthanide element plays an important role in the formation of the dense random packing structure of (Fe, Co)–B in these alloys.[6] The supercooled liquid region ΔT_x of Fe–Co–Ln–B glassy alloys are about 40–63 K.[7] The bulk glassy rods with diameters of 0.5 and 0.75 mm for $Fe_{62}Co_{9.5}Nd_3Dy_{0.5}B_{25}$ were producted by a copper mold casting method.[5] However, it is necessary to search for an additional element which increases the glass-formation ability of Fe–Co–Ln–B glassy alloys to improve their thermal stability of supercooled liquid. In this article, we examine the effects of the additional TM (TM=transition metals) on the glass-formation ability and magnetic properties of $Fe_{62}Co_{9.5}Nd_3Dy_{0.5}B_{25}$ glassy alloy.

II. EXPERIMENTAL PROCEDURE

Alloy ingots of $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ (TM=V, Nb, Ta, Cr, Mo, and W) were prepared by arc melting the mixtures of pure metals and B crystal in an argon atmosphere. Then the ingots were crushed into small pieces in order to place them into a quartz crucible for melt spinning and copper mold casting. The nozzle size of the crucible was about 0.5 mm in diameter. Ribbons were produced by melt spinning with a wheel speed of 30 m/s in an argon atmosphere. Bulk samples of rod shape were produced by injection casting of the molten alloy into copper molds. The struc-

[a)]Author to whom correspondence should be addressed; electronic mail: wzhang@sendai.jst.go.jp

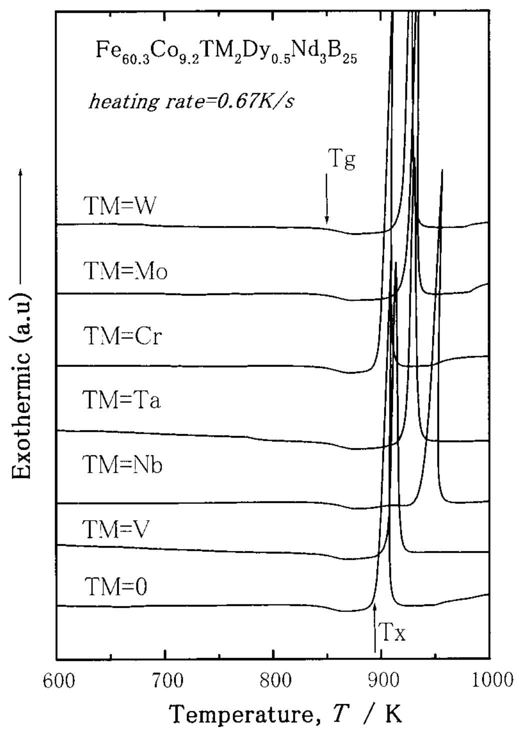

FIG. 1. DSC curves of melt-spun $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloys.

TABLE I. Magnetic properties and thermal stability of the melt-spun $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ ribbons.

Composition	I_s(T)	H_c(A/m)	T_g(K)	T_x(K)	ΔT_x(K)
TM=Cr	1.16	3.85	843	898	55
TM=Mo	1.15	4.98	843	924	81
TM=W	1.14	3.98	845	921	76
TM=V	1.13	4.52	847	907	60
TM=Nb	1.15	4.78	850	937	87
TM=Ta	1.19	3.98	850	924	74
TM=0	1.37	4.78	842	897	55

ture of samples was examined by x-ray diffraction (Cu $K\alpha$) and transmission electron microscopy. Thermal stability was examined by differential scanning calorimeter (DSC) and differential thermal analysis in an argon atmosphere at a heating rate of 0.67 and 0.033 K/s, respectively. The saturation magnetization was measured at room temperature by a vibrating sample magnetometer with a maximum applied magnetic field of 670 kA/m. The coercive force was measured with a B–H loop tracer.

III. RESULTS AND DISCUSSION

Figure 1 shows DSC curves of the melt-spun $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ ribbons, where T_g and T_x are glass transition temperature and crystallization temperature, respectively. As seen in Fig. 1, the glass transition temperature T_g remains almost unchanged and the crystallization temperature T_x shifts toward higher temperature by the addition of the TM elements. All the glassy alloys crystallize through a single stage with an exothermic heat of 5.26–7.49 kJ/mol. Based on the DSC curves shown in Fig. 1, $\Delta T_x(=T_x-T_g)$ values are plotted as a function of elements TM. From Fig. 2, it is observed that ΔT_x increases significantly from 55 to 87 K with the replacement of 2 at. % Fe and Co by elements of Nd, Ta, Mo, and W. Figure 2 also shows the data on the onset temperature of solidification T_m obtained from the continuous cooling curves of the $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ molten alloys. The T_m decreases with the replacement of 2 at. % Fe and Co by Mo, W, Nb, and Ta. Obviously, the addition of TM (=Mo, W, Nb, and Ta) elements into the Fe–Co–Ln–B alloy is very useful for the suppression of crystallization. The results can be explained by the three empirical rules for the increase in the thermal stability of the supercooled liquid against crystallization.[3] The addition of TM (=Mo, W, Nb, and Ta) elements causes the more sequential change in atomic size on the order of Nd>Dy>TM(TM=Mo,W,Nb,and Ta)>Fe >Co>B because atomic diameters of TM (=Mo, W, Nb, and Ta) are between those of Fe, Co, and Nd. The addition of TM (=Mo, W, Nb, and Ta) increases the degree of dense

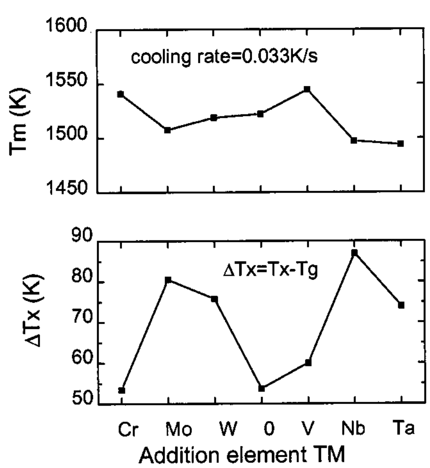

FIG. 2. $\Delta T_x(=T_x-T_g)$ and T_m as a function of the transition metals TM for the glassy $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloys.

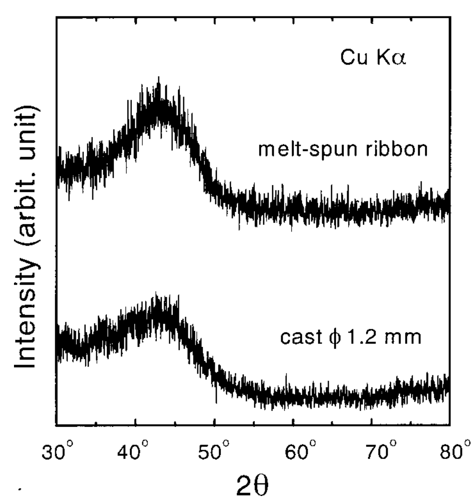

FIG. 3. X-ray diffraction pattern of the cast glassy $Fe_{60.3}Co_{9.2}Nb_2Nd_3Dy_{0.5}B_{25}$ cylinder with a diameter of 1.2 mm. The data of the melt-spun glassy ribbon with the same composition are also shown for comparison.

random packing fraction and difficulty for crystallization. It is also seen that the characteristic of the transition metals can affect the thermal stability against crystallization by comparing the addition of Nb with Ta. Otherwise, the thermal stability against crystallization is not slightly improved with the addition of Cr and V elements because the atomic diameters of Cr and V are close to those of Fe and Co. The wider ΔT_x and lower T_m for $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ (TM=Nb, W, Mo, and Ta) indicate that the present glassy alloys have a much higher glass-formation ability. The maximum T_x and the lowest T_m are obtained for $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$. They are 87 and 1497 K, respectively. These data are better than the best values for the previously reported glassy Fe–Co–Ln–B alloys.[4–7]

Table I summarizes the magnetic properties and thermal stability of the melt-spun $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ ribbons. The $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloys have better saturation magnetization of 1.13–1.19 T and lower coercive force of 3.85–4.98 A/m. The saturation magnetization of $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ is slightly lower than that of $Fe_{62}Co_{9.5}Nd_3Dy_{0.5}B_{25}$ because nonferromagnetic elements of TM are added instead of ferromagnetic elements of Fe and Co.

The cast $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloy in a cylinder form with a diameter of 1.2 mm was produced. It is confirmed that the cast alloy consists of a glassy single phase from x-ray diffraction patterns and DSC curve. Figure 3 shows x-ray diffraction patterns of the cast alloy and the melt-spun glassy ribbon. No distinct difference in x-ray diffraction patterns is seen between the bulk sample and the melt-spun glassy ribbon. The saturation magnetization of the cast alloy is 1.15 T.

IV. SUMMARY

We have examined the effects of the addition transition metals TM(TM=V, Nb, Ta, Cr, Mo, and W) on the glass-formation ability and magnetic properties for the glassy $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloy. The addition of TM (=Nb, Ta, Mo, and W) significantly increases crystallization temperature T_x and decreases the onset temperature of solidification T_m, leading to a significant increase in the thermal stability against crystallization for $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$. ΔT_x increases up to 87 K at 2 at. % TM from 55 K at 0 at. % TM (=Nb, Ta, Mo, and W). Also no distinct changes in T_g, T_x, and ΔT_x are seen for the addition of Cr and V. The saturation magnetization I_s and coercive force H_c of the glassy $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloys are 1.13–1.19 T and 3.85–4.98 A/m, respectively. The saturation magnetization decreases slightly by the addition of TM. Bulk glassy $Fe_{60.3}Co_{9.2}TM_2Nd_3Dy_{0.5}B_{25}$ alloy with a diameter of 1.2 mm was produced by copper mold casting.

[1] A. Makino, A. Inoue, and T. Mizushima, Mater. Trans., JIM **41**, 1471 (2000).
[2] R. Hasegawa, J. Magn. Magn. Mater. **215–216**, 240 (2000).
[3] A. Inoue, *Bulk Amorphous Alloys* (Trans Tech Publishers, Zurich, 1998).
[4] A. Inoue and W. Zhang, J. Appl. Phys. **85**, 4491 (1999).
[5] W. Zhang and A. Inoue, Mater. Trans., JIM **41**, 1679 (2000).
[6] M. Imafuku, K. Yaoita, S. Sato, W. Zhang, A. Inoue, and Y. Waseda, Mater. Sci. Eng., A **304–306**, 660 (2001).
[7] W. Zhang and A. Inoue, Mater. Trans., JIM **42**, 1142 (2001).